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Flux Penetration Effects in High-T_c SQUIDs*

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Abstract

Two experiments pertaining to the effects of intergranular flux penetration in high-T_c yttrium–barium–copper oxide (YBCO) SQUIDs are described. The first is a direct measurement of the flux noise of bulk YBCO exposed to the earth's magnetic field, and the second involves the fabrication and testing of break junction d.c. SQUIDs. Implications of a number of undesirable effects seen in these experiments are discussed.

1. Introduction

Work at the CSIRO Division of Applied Physics on SQUIDs arose out of the development of the Josephson voltage standard in the early 1970s (Harvey *et al.* 1972). During the 1970s several types of niobium point-contact r.f. SQUIDs were developed for various metrological purposes. More recently research has spread in several new directions. A facility for producing thin-film niobium–lead alloy d.c. SQUIDs has been established (Sloggett *et al.* 1988) and SQUIDs for use in neuromagnetic measurements are under development. At the same time extensive work has been undertaken on yttrium–barium–copper oxide (YBCO) SQUIDs. Harvey *et al.* (1988) reported the performance of both single-hole and two-hole (gradiometer) break junction r.f. SQUIDs operating at 77 K. A substantial program of research is aimed at producing YBCO thin films suitable for SQUID fabrication, amongst other applications (Smith *et al.* 1989; present issue p. 431).

In this paper we report a direct measurement of the low-frequency magnetic flux noise of bulk YBCO. We also describe our partly successful attempts to operate a break junction YBCO d.c. SQUID. Several types of undesirable behaviour are noted, most or all of which are attributable to flux penetration and movement in intergranular regions of YBCO. The implications of these experiments for the likely performance of future optimised thin-film 77 K SQUIDs are discussed.

2. Measurement of Flux Noise in YBCO

The first high-T_c SQUIDs on which noise measurements were made were of the r.f. type, made from bulk YBCO with either intergranular junctions

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(Colclough *et al.* 1987; Pegrum *et al.* 1987), break junctions (Zimmerman *et al.* 1987; Harvey *et al.* 1988), or eroded constriction junctions (Harrop *et al.* 1988). These have generally shown levels of white noise not much higher than for liquid helium SQUIDs (in the range 10^{-3}–10^{-4} $\phi_0\,\mathrm{Hz}^{-1/2}$, where ϕ_0 is the flux quantum, 2×10^{-15} Wb), but rather high levels of low frequency, or $1/f$, noise. Harvey *et al.* (1988) found that the $1/f$ noise region extended up to about 100 Hz, well into the frequency range of interest for major applications such as biomagnetism and geomagnetism. Noise measurements have also been reported for a number of thin-film YBCO d.c. SQUIDs, though generally at temperatures below 77 K (Takeuchi *et al.* 1988; Matsuda and Kuriki 1988). Sandstrom *et al.* (1988) operated a thin-film YBCO d.c. SQUID at 77 K and obtained a measured sensitivity, dominated by $1/f$ noise, of 3×10^{-4} $\phi_0\,\mathrm{Hz}^{-1/2}$ at 20 Hz. The same group has recently reported thin-film thallium–barium–calcium–copper oxide (TBCCO) SQUIDs with the best noise performance reported to date: below 10^{-4} $\phi_0\,\mathrm{Hz}^{-1/2}$ at 10 Hz (Koch *et al.* 1989).

In low temperature SQUIDs $1/f$ noise is a widely observed phenomenon, and a recent study has identified four different types of noise (Wellstood *et al.* 1987). One of these is a material-dependent flux noise, thought to be associated with the movement of flux vortices trapped within the material forming the SQUID loop. The three other sources of noise are various processes occurring at the SQUID Josephson junctions, such as critical current fluctuations. The experiment described here was undertaken to distinguish between these two types of mechanism for the high level of $1/f$ noise seen in 77 K YBCO SQUIDs.

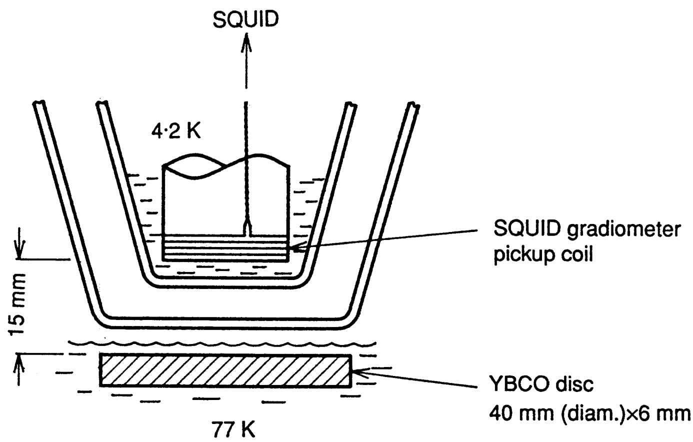


Fig. 1. Apparatus for measurement of YBCO flux noise.

Our apparatus (Fig. 1) consisted of a 4·2 K SQUID gradiometer system (CTF Systems Inc 3rd order) with a magnetic field sensitivity of 37 $\mathrm{fT\,Hz}^{-1/2}$ referred to its lower pickup coil. A YBCO disc 40 mm in diameter and 6 mm thick was placed in liquid nitrogen at a distance of about 15 mm from the gradiometer pickup coil. The material was prepared by wet ball milling appropriate amounts of Y_2O_3, $BaCO_3$ and CuO, followed by reaction of the dried powder at 900–920°C. This precursor material was packed in an alumina dish and fired at 950°C for 14 hours, followed by slow cooling in air. The sintered disc had an approximate density of $4\cdot1\ \mathrm{g\,cm}^{-3}$ (64% of theoretical maximum) and was

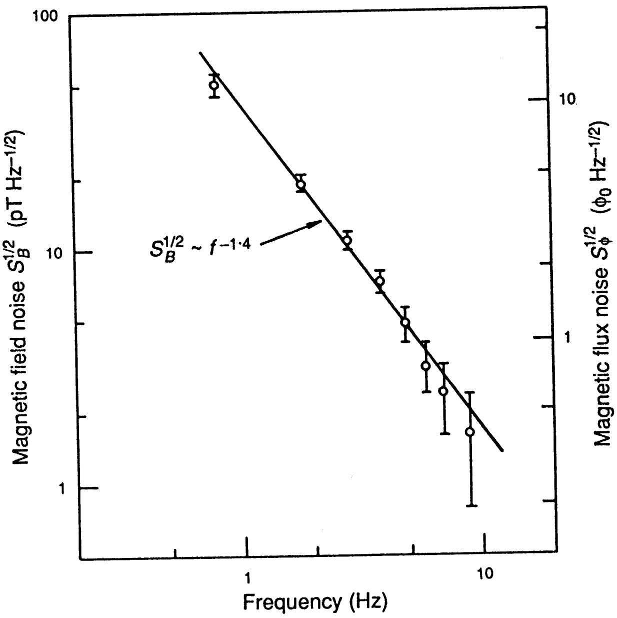


Fig. 2. Measured low-frequency noise in YBCO.

shown to be phase pure by X-ray diffraction measurements. Care was taken to eliminate vibration of the disc in the earth's magnetic field as a possible source of noise.

Measured results (Fig. 2) show a large level of magnetic field or flux noise, varying approximately as $f^{-1\cdot4}$ over the frequency range studied. Expressed as an equivalent flux noise the noise level was about 10 $\phi_0\,\mathrm{Hz}^{-1/2}$ at 1 Hz. This is at least two orders of magnitude higher than the flux noise levels which have been observed in bulk YBCO SQUIDs. Direct comparison is complicated by differences in the dimensions and separation of the noise-generating sample and the pickup loop (these are one and the same in the case of an YBCO SQUID), and by possible differences in material quality (e.g. a porous sample might be expected to exhibit different flux dynamics to a dense sample). However, probably the principal reason for the high level of flux noise observed in the present measurements is that they, unlike any of those made on SQUIDs, were undertaken in an unshielded magnetic environment; flux motion noise can be expected to increase with increasing flux density. This is of some practical significance since, for many applications, superconducting flux transformers, if not the SQUID itself, must be exposed to the earth's magnetic field. Our results show that bulk YBCO in such an environment is a very noisy material. The level of noise, if duplicated in a 77 K SQUID magnetometer system, would be sufficient to preclude its use in any major low-frequency application.

Our results may be compared with those of Ferrari *et al.* (1988), who used a 4·2 K SQUID system to measure the magnetic flux noise of YBCO thin films. They found that the $1/f$ noise varied with film quality (the best film was strongly oriented with its c-axis normal to the substrate and had a high critical

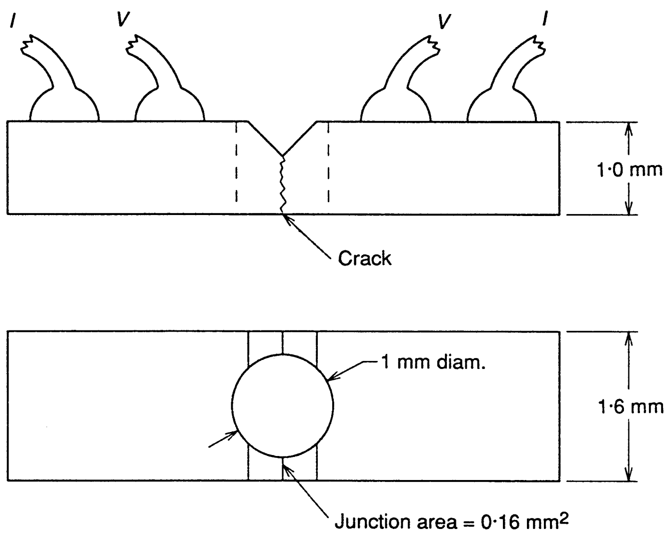


Fig. 3. Design for break-junction YBCO d.c. SQUID.

current density) and increased with temperature, especially as T approached T_c. Their measurements were made in a well shielded environment and the measured noise level at 1 Hz and 77 K was below $10^{-2}\ \phi_0\,\mathrm{Hz}^{-1/2}$ for the two best films.

3. 77 K Break-junction DC SQUIDs

Several break-junction d.c. SQUIDs have been constructed from bulk YBCO and have been operated with varying degrees of success. Fig. 3 shows one design; others differ slightly in dimensions. The objectives were to achieve a structure with low inductance and with two junction regions of small, well-matched cross-sectional area. The material used was pressed and sintered granular YBCO prepared as described by Harvey *et al.* (1988). The two ends of the devices were cemented to opposing jaws of brass structures with a 'scissors' geometry providing a large mechanical advantage. The brass structures were provided with screw adjustments allowing the SQUID to be slowly placed in tension, inducing cracks in the junction regions. Junction formation was carried out in liquid nitrogen and the device current–voltage characteristics monitored during adjustment. Small coils placed within the central hole or adjacent to it allowed a variable magnetic field to be applied to the SQUID.

Fig. 4 shows the current–voltage characteristics of one device at 77 K after crack adjustment had reduced the critical current by at least one order of magnitude. The two superimposed traces correspond to two different applied fields, and strong modulation of the critical current (of order 100 μA p–p) is evident. The current modulation is about an order of magnitude stronger than expected for a SQUID of inductance of order 10^{-10} H. When the device is biased at a suitable d.c. current it is found that its output voltage exhibits a correspondingly strong, but non-periodic, field modulation (Fig. 5*a*). The

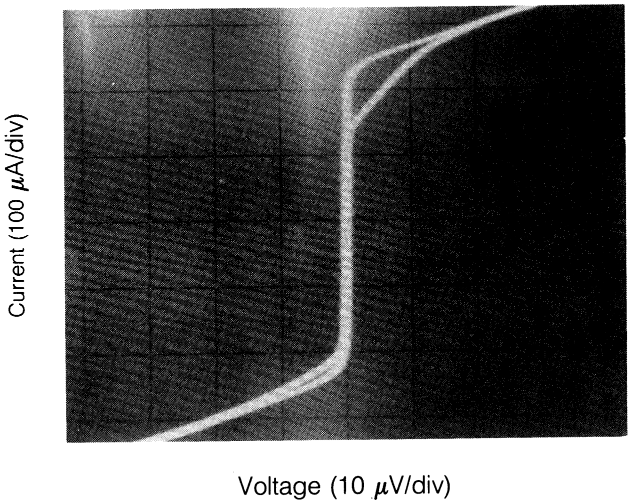


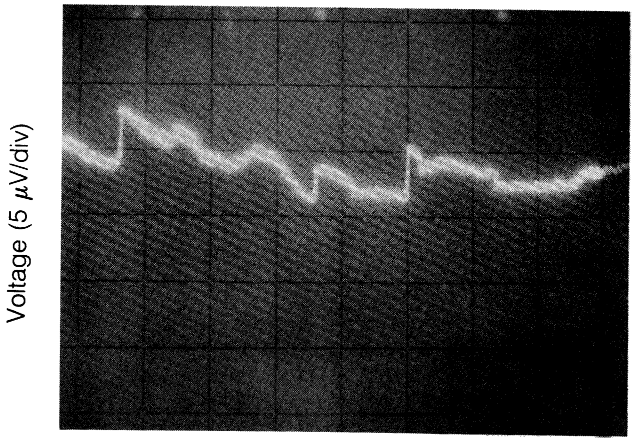
Fig. 4. SQUID current–voltage characteristics.

voltage–field characteristics were hysteretic, the characteristics for increasing field differing substantially from that for decreasing field. The large amplitude and aperiodicity of the field modulation indicates that the dominant response of this device is not due to flux passing through the central hole, but more likely to quantum-interferometric behaviour in multiple low-inductance intergranular loops. The true SQUID response of the best of our devices was observable as a low-amplitude (about 1 μV p–p) voltage response superimposed on the large amplitude fluctuations (Fig. 5*b*). This response has the appearance of a series of sinusoidal segments interrupted by cusps and discontinuities. The explanation for these interruptions of the normal periodic interferometric response is unclear, although similar behaviour has been observed elsewhere in thin-film YBCO SQUIDs (Takeuchi *et al.* 1988).

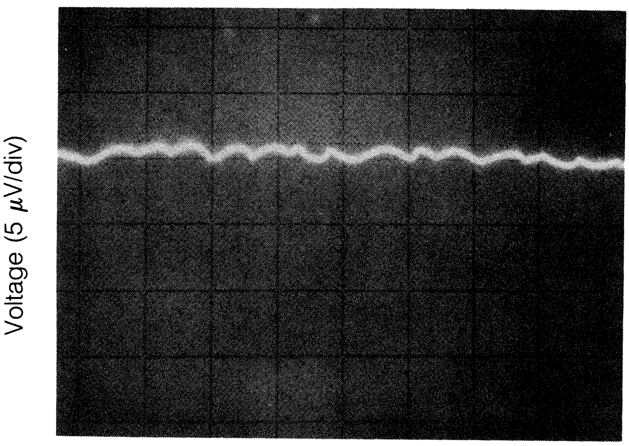
In summary, although some of our devices have shown true d.c. SQUID response, various deficiencies in their voltage–field characteristics make these devices unsatisfactory for practical use. Because of these shortcomings, we have not attempted to operate a device in a flux-locked loop or to measure its noise. However, an estimate may be made of the white noise of the device by applying standard d.c. SQUID theory (Clarke 1980) and using measured values of the SQUID resistance and voltage–flux transfer function. We estimate the white noise of the device illustrated in Figs 4 and 5 to be about $2\cdot5\times10^{-5}\,\phi_0\,\mathrm{Hz}^{-1/2}$, and its energy resolution, assuming an inductance of about 10^{-10} H, to be $2\cdot5\times10^{-29}\,\mathrm{J\,Hz}^{-1}$.

4. Discussion

Our experiments on noise and d.c. SQUID fabrication reveal several problems for YBCO SQUIDs which must be overcome before practical 77 K sensors can be realised. Firstly, bulk granular YBCO is an intrinsic generator of low-frequency magnetic noise, the intensity of which appears to be enhanced on exposure to the earth's magnetic field. Secondly, SQUIDs made from this material exhibit



(*a*) Magnetic field × 1 (arb. units)



(*b*) Magnetic field × 10 (arb. units)

Fig. 5. SQUID voltage–field characteristics: (*a*) and (*b*) differ by a factor of 10 in field scale.

several undesirable features in their response to applied magnetic fields. The anticipated periodic quantum interferometric response is observed, but it is interrupted by numerous cusps and discontinuities and is dominated by large-amplitude interferometric response which is both aperiodic and hysteretic.

Although much theoretical detail remains to be filled in, there is little doubt that some, if not all, of the features observed are attributable to the penetration and movement of flux vortices in the intergranular regions of bulk YBCO. Müller *et al.* (1989*a*; 1989*b*, present issue p. 413) found that the pinning force density for vortices in the intergranular regions of a bulk YBCO sample at 77 K was about 116 $\mathrm{T\,A\,m^{-2}}$, or more than seven orders of magnitude weaker than for NbTi at 4·2 K. The material used in these experiments was prepared in the same way as that used in our SQUIDs and had a similar density (about 87% of theoretical maximum). A magnetic field of less than 10^{-4} T is sufficient to cause flux to penetrate between grains in samples of a few mm thickness. The movement of weakly pinned flux (flux jumps) when the applied field is varied accounts for the hysteresis of SQUID characteristics, and possibly also for their cusps and discontinuities, as suggested by Takeuchi *et al.* (1988). Lattice vibrations provide another possible source of energy for unpinning of flux vortices, and thermally activated flux motion is the probable cause of the high $1/f$ noise of YBCO material and SQUIDs. The rapid rise in the $1/f$ noise level of YBCO films as the temperature approaches T_c (Ferrari *et al.* 1988) is consistent with the measurements of Ricketts *et al.* (1989) showing that the pinning forces decrease monotonically to zero over this range. Finally, the spurious quantum interference effects observed in our SQUIDs are also related to flux penetration into intergranular Josephson junction networks. Some r.f. SQUIDs which directly exploit this property have been demonstrated (Colclough *et al.* 1987; Pegrum *et al.* 1987).

It appears that significant improvements in material properties are necessary for practical 77 K SQUID operation. Material with significantly enhanced intergranular pinning forces, or else few or no grain boundaries, would appear to be required. This is most likely to be achieved in thin films. New materials, particularly with higher T_c values, may help. Koch *et al.* (1989) have recently described thin-film TBCCO SQUIDs having much lower noise than any YBCO SQUID reported to date, although low-frequency noise still apparently dominates the sensitivity of these devices, and hysteresis in the response to applied fields remains a problem.

Provided the low-frequency noise and other flux penetration problems can be overcome, there are grounds for optimism about the performance of 77 K SQUIDs. An energy resolution better than 10^{-28} J Hz^{-1} is estimated for our break-junction d.c. SQUIDs operating in the white noise frequency range and indeed has been measured in the TBCCO SQUIDs of Koch *et al.* (1989) at 10 Hz. Such a sensitivity, extending down to sufficiently low frequencies in a SQUID of practical design, would be adequate for many potential applications (e.g. geomagnetism, magnetocardiology). It appears for the present that 4·2 K SQUIDs will continue to be used for the most demanding applications such as magnetoencephalography.

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